

Evaluation of uranium thorium and plutonium thorium fuel cycles in a very high temperature hybrid system

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ABSTRACT

In recent times, there is a renewed and additional interest in thorium because of its interesting benefits. Thorium fuel cycle is an attractive way to produce long term nuclear energy with low radiotoxicity waste. In addition, the transition to thorium could be done through the incineration of weapons grade plutonium or civilian plutonium. Th-based fuel cycles have intrinsic proliferation-resistance and thorium is 3–4 times more abundant than uranium. Therefore, thorium fuels can complement uranium fuels and ensure long term sustainability of nuclear power.

In this paper, the main advantages of the use of fuel cycles based on uranium-thorium and plutonium-thorium fuel mixtures are evaluated in a hybrid system to reach the deep burn of the fuel. To reach this goal, the preliminary conceptual design of a hybrid system composed of a critical reactor and two Accelerated Driven Systems, of the type of very high temperature pebble-bed systems, moderated by graphite and cooled by gas, is analyzed.

Uranium-thorium and plutonium-thorium once-through and two stages fuel cycles are evaluated. Several parameters describing fuel behaviour and minor actinide stockpile are compared for the analyzed cycles.

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1. Introduction

Deep burn transmutation in Very High Temperature Reactors (VHTR) is based on using the neutron thermal spectrum to reach a high fuel burnup. Simplified comparisons made between capture and fission cross sections of fast and thermal neutrons concluded that transmutation using thermal reactors is not feasible. Nevertheless, more detailed studies carried out later proved that thermal neutrons are capable to transmute Minor Actinides (MA) in appropriate conditions (Venneri et al., 2001).

In the deep burn concept, transmutation of long lived wastes composed of transuranic elements from the nuclear reactors and almost the whole destruction of the materials useful for the nuclear weapons fabrication (plutonium isotopes), is obtained with only once-through reprocessing cycle (Baxter and Rodriguez, 2001).

One of the key benefits of the proposed system in this paper is the utilization of the fuel confined into TRISO (Tri-structural isotropic) particles. Graphite coated fuel particles present some attractive advantages for deep burn transmutation as they have

high resistance to irradiation damages, mechanical stress, and have high melting points, enabling to reach a high fuel burnup (Maki et al., 2007). Such performance is central to the deep burn transmutation concept (Fokuda et al., 1995). The different TRISO particle's layers constitute excellent fission product and radionuclide barriers in the geological repository (US Nuclear Regulatory Commission, 2004). Ceramic fuel particles also keep impenetrable to humidity longer than current metallic containers destined to preserve the spent fuel of conventional nuclear reactors. These features make TRISO particles a robust and attractive waste container.

VHTRs belong to next nuclear plants generation, the Generation IV (U.S. DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, 2002). VHTRs are expected to have attractive features like low electricity generation costs and short construction periods. The development of this technology is based on the experience from High Temperature Gas Reactors (HTGRs) like Dragon Peach Bottom, from England, and German Arbeitsgemeinschaft Versuchsreaktor (AVR) and Thorium High Temperature Reactor (THTR), from Germany. The above mentioned are experimental reactors that were built in the 60's to demonstrate their viability for electricity and heat cogeneration. They enabled to reach high coolant temperatures at the core's outlet. Current

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projects, i.e. HTTR-2000, from Japan and HTR-10, from China, which became operatives in the years 2000 and 2003 respectively, enable to reach coolant outlet temperatures higher than 950°C (Talamo and Gudowski, 2005).

Taking advantage of the high coolant's outlet temperatures in a VHTR, it is possible to produce hydrogen from water and heat utilizing the Iodine–Sulfur thermochemical process, or from water, heat and natural gas applying steam reforming. Electricity production with a high thermodynamic efficiency is also possible in a direct cycle with a gas turbine. The heat extracted from the reactor is also useful in refineries, petrochemical and metallurgic industries (Abánades and García, 2009).

Because of its constructive characteristics, TRISO fuel can reach a high burnup as mentioned above. It could work in a VHTR and later in a Very High Temperature Accelerator Driven System (ADS). This strategy would enable to reach the allowed high burnup for this kind of fuel with no structure damages due to irradiation or high temperatures.

An ADS is composed of a heavy particles accelerator, where high energy protons are generated. Protons react with a spallation target composed of heavy materials to produce neutrons. The spallation target is located at the centre of a subcritical core which contains the nuclear fuel. The nuclear fuel may be composed of traditional nuclear fuel, transuranic elements, long lived fission products to transmute them into short lived isotopes, or uranium-thorium (U + Th) and plutonium-thorium (Pu + Th) mixtures, as in the present study. Fissile elements are disposed in a way that nuclear chain reaction cannot be sustained without a neutron source (in this case are the neutrons from spallation reactions). Neutrons and charged particles are generated from spallation reactions when the accelerated protons hit the target. These neutrons are tightly linked to the kinetic behaviour of the system, like delayed neutrons in critical reactors. Therefore, the low fraction of delayed neutrons of the spent fuel isotopes loses importance in the chain reaction control, which is determined by the neutron source and the accelerator.

Neutrons produced from spallation reactions in the target and multiplied by fission neutrons in the nuclear fuel, can transmute transuranic elements and long lived fission products into stable elements. As a result of fission reactions that take place in the transmutation processes, large quantities of heat are produced and can be transformed into electricity.

The calculation methods used in this work are based on probabilistic computational modelling. The principal tool for particle transport and fuel burnup calculations was MCNPX code, version 2.6e (McKinney et al., 2007). This software was created in 1994 by Los Alamos National Laboratory. It is based on the Monte Carlo probabilistic method for computational modelling of time dependent transport of many kinds of particles, with a wide range of energies, in much different geometries. Version 2.6e incorporates new capabilities with respect to previous versions. Some of them were used in the present study, such as CINDER90 code for fuel burnup calculation (Wilson et al., 1999). The available library in XSDIR, ENDF/B VI.2, was used.

In the present work, we analize the feasibility of once-through cycles based on U + Th or Pu + Th fuel mixtures, with two stages: the first in a very high temperature pebble-bed graphite-gas critical reactor, and the second in two ADSs with similar characteristics of the reactor. To reach this goal, we propose a preliminary conceptual design of the hybrid system composed of the mentioned critical reactor and the two ADSs. We evaluated the advantages of the proposed fuel cycles and compared them with traditional cycles in terms of those parameters that describe the behaviour of fissile fuel and MA stockpile.

The characteristics and advantages of thorium based fuel cycles are introduced in Section 2. In Section 3 a once-through and

two-stages (a VHTR and two ADSs) fuel cycle in a VHTR hybrid system is shown and described. The conceptual design of a very high temperature hybrid system for a U + Th deep burn cycle is made in Section 4. Some of the analyzed items are: the optimization of the physical parameters, fuel burnup, variation of the isotopic composition and the radiotoxicity of the long lived wastes. By the end of the section a comparison between a U + Th cycle and an equivalent uranium cycle is presented. In Section 5 the possibilities of Pu + Th cycles in the very high temperature hybrid system are studied. Finally, some conclusions are discussed in Section 6.

2. Uranium + thorium cycle

In uranium-thorium fuel cycle, reactor's criticality is mainly sustained by three isotopes: U^{235} , which represents a certain percent of the fresh uranium; U^{233} , which is produced by transmutation of the fertile isotope Th^{232} , and Pu^{239} , produced by transmutation of the fertile isotope U^{238} . In addition, Pu^{241} , formed by neutronic capture of Pu^{240} , modestly contributes to the energy released by fission during the fuel cycle, but its contribution to the final inventory of fissile isotopes is poor because of its short half life, as it is converted almost completely into Am^{241} during the cooling time of the wasted fuel.

In order to compensate the U^{235} depletion by means of U^{233} and Pu^{239} breeding, the amount of fertile nuclides must be greater than the amount of U^{235} due to the small capture cross section of the fertile nuclides in the neutron thermal energy range compared to the capture cross section of the U^{235} . At the same time, the amount of U^{235} must be large enough to set the criticality of the reactor (Talamo and Gudowski, 2005).

In nature, thorium exists as a natural radioactive metallic element. The general certain about thorium stockpile and allocation is quite limited because the global demand of thorium has been rather insignificant.

A number of thorium-based fuel cycle benefits follow (International Atomic Energy Agency, 2005):

- It is 3–4 times as abundant as uranium, is widely distributed in the earth crust and is easily commercially exploitable.
- Thorium fuel cycle is a very attractive way to produce long term nuclear energy with low level radiotoxicity waste. In addition, the transition to thorium could be done through the incineration of weapons grade plutonium or civilian plutonium.
- The absorption cross section of Th^{232} for thermal neutrons (7.4 b) is nearly three times that of U^{238} (2.7 b). Hence, the conversion of Th^{232} into U^{233} is theoretically higher than the currently used transformation of U^{238} into Pu^{239} . Therefore, thorium could be a better fertile material than uranium in a thermal neutron spectrum.
- For the fissile nuclide U^{233} , the number of fast neutrons produced per thermal neutron absorbed is greater than that for U^{235} and Pu^{239} . Thus, in contrast to the U^{238} – Pu^{239} cycle, in which breeding can be obtained only with fast neutron spectrum, in the Th^{232} – U^{233} fuel cycle breeding can be obtained with fast, epithermal or thermal spectra.
- The U + Th fuel cycle produces a lower amount of Pu and MA (Np, Am and Cm) than the U + Pu fuel cycle, which contributes to minimize the radiotoxicity associated to the spent fuel. Consequently, a thorium based fuel cycle produces less hazardous waste than the U–Pu fuel cycle used in current LWRs.

Some challenges associated to the use of thorium based fuels are under investigation (International Atomic Energy Agency, 2005).

3. Once-through and two-stages fuel cycle in a very high temperature hybrid system

In the present work, the performing of once-through and two-stages fuel cycles based on U + Th and Pu + Th fuel mixtures, in a very high temperature hybrid system, reaching a deep fuel burnup, is proposed. In the first case, the fuel is a mixture of U + Th composed of 60% of uranium and 40% of Th²³² by weight, and in the second case the fuel is a mixture of Pu²³⁹ and Th²³². The proportions for the used mixtures of Pu–Th will be explained in Section 5. The Fig. 1 shows the different steps of once-through and two-stages fuel cycle in Very High Temperature Advanced Nuclear Systems.

3.1. The reactor

The proposed reactor is a VHTR designed for the deep burn of the fuel. Its fuel is composed of enriched uranium and thorium in the first case and by Pu²³⁹ and thorium in the second case. The core is a pebbled-bed type cooled by helium and moderated by graphite. This kind of reactor reaches a high energy conversion efficiency because of the greater thermodynamic efficiency than present gas cycles compared to the steam cycles commonly used in LWRs (McKinney Gregg et al., 2007). VHTRs are good in once-through fuel cycles and allow obtaining high coolant outlet temperatures.

In the conceptual design of the proposed once-through and two-stages fuel cycle, the following reactor core's characteristics have been considered (Table 1). The cylindrical core is surrounded by a graphite reflector.

3.2. The accelerator driven systems

As a second stage, the fuel is extracted from the reactor and loaded in two ADSs, in order to reach the deep burn of the U + Th and Pu + Th mixtures. The ADSs parameters were chosen based on a conceptual design reported in Abánades and García (2011), called "Transmutation Advanced Device for Sustainable Energy Applications", which will be called from this point as "the reference ADS" (Table 2). ADSs geometry was designed in a way that exactly the half amount of the reactor pebbles were placed on each ADS. The

Table 1
Parameters of the Reactor's core.

Thermal power (MW _{th})	200
Core's Height (m)	2.94
Core's Diameter (m)	4.17
Average Power Density (MW/m ³)	5
Volume (m ³)	40.24
Number of fuel elements	263465

suggested accelerator is a cyclotron with six separated sectors and four accelerating cavities enabling to reach energies of 1 GeV with a proton beam intensity of 10 mA. The spallation target is based on eutectic lead bismuth mixture. The general features of the target were studied in detail in Abánades and Pérez–Navarro (2007).

The core of the ADSs is a cylinder containing the fuel elements. Its central part is occupied by the spallation target which is the neutron source. Core is filled with the graphite pebbles extracted from the reactor; therefore, this fuel has been exposed to neutron radiation and contains fission products. The reactor and both ADSs are continuously refuelled. For simulation purposes, refuelling was divided into ten partial recharges in which one tenth of the volume is refuelled each one tenth part of the cycle.

Gas helium was chosen as coolant, in reactor and ADSs, despite it is more expensive than carbon dioxide or nitrogen, because it has some advantageous features: it does not moderate neutrons, whence neutron spectrum will be only determined by the fuel design, and on the other hand, there will be no reactivity coefficient due to coolant temperature because a helium density variation will not produce an appreciable increase of its absorption and elastic neutron cross sections. Also it is chemically inert and does not produce graphite oxidation.

4. Conceptual design of a very high temperature hybrid system for a once-through and two-stages U + Th deep burn cycle

4.1. Physical parameters optimization

In the first cycle's stage, the fuel is burned during 200 days in the VHTR under a power of 200 MW_{th}. When the criticality of the

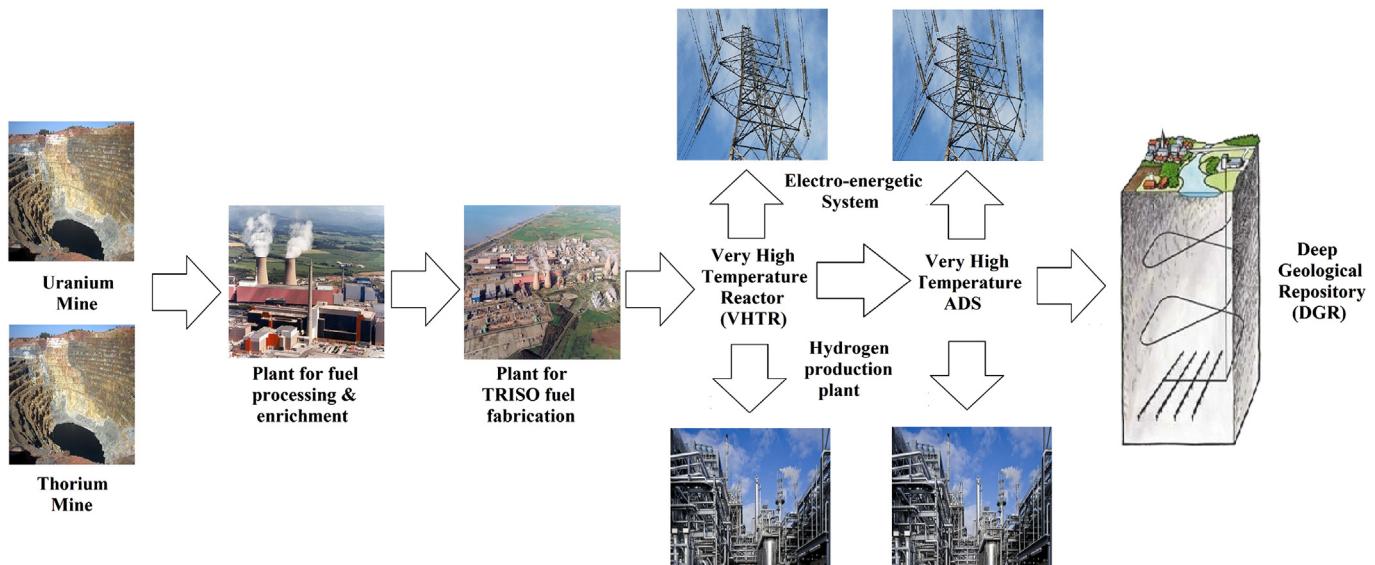


Fig. 1. Once-through and two-stages fuel cycle in very high temperature advanced nuclear systems.

Table 2

Main parameters of the proposed ADSs and the reference ADS.

Parameter	Ref. ADS	ADSs
Accelerator power (MW)	10	10
Proton beam energy (MeV)	1000	1000
Proton beam intensity (mA)	10	10
Core's inner radius (cm)	15.5	15.5
Core's outer radius (cm)	125.75	148.42
Height (cm)	293.94	293.94
Total volume (m ³)	14.38	20.12
Fuel volume (m ³)	6.16	8.62
Fuel mass (kg)	124.5	510.1
Number of pebbles	94092	131730
Packing fraction	0.74	0.74
K _{eff}	0.94	0.75
Thermal power (MW _{th})	100	60
Average power density (W/cm ³)	7	3
Radial reflector thickness (cm)	60	180

system cannot be maintained, the fuel pebbles are moved to two ADSs, in which they will remain during 110 days under a power of 60 MW_{th}.

The core's dimensions design was performed assuming a working power of 200 MW_{th} and an average power density of 5 MW/m³, since this value is under the accepted value 7 MW/m³, corresponding to this type of reactors. A packing fraction of 0.74 was chosen for the reactor's core, as this is the highest possible packing fraction in an infinite ordered face-centered pebble bed configuration (Tian, 2007).

The fuel burnup in the reactor was modelled in 10 time steps of 20 days. At the beginning of each step, the lowest layer of fuel elements is extracted from the reactor; then, the extracted layer is divided into two equal parts and each part is introduced on the upper layer of each ADS core. Every layer of fuel of the reactor's core drops one level and fresh fuel is introduced on the upper layer.

In order to obtain the optimal reflector thickness, the effective multiplication factor (K_{eff}) dependence on different reflector thickness values was studied. Results shown in Fig. 2 indicate that the maximum reflector saving is obtained with a radial reflector thickness of 180 cm. Hence, radial and axial graphite reflector thicknesses of 180 cm and 100 cm respectively, were considered for the cylindrical core. The control rods and the gas coolant supplier ducts are proposed to be located in the radial reflector. The results obtained in the studies of the reactor reflector design were used for the ADSs cores design.

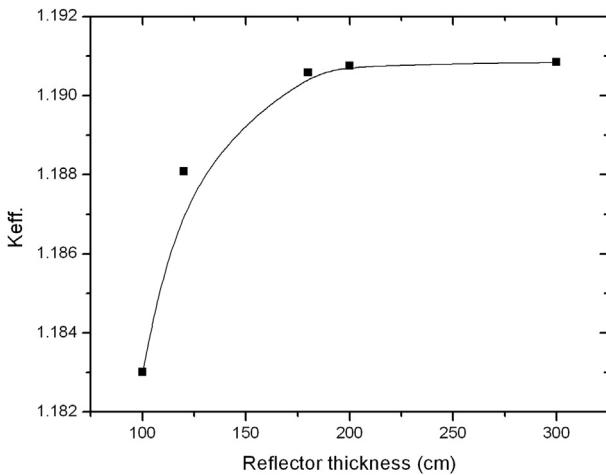
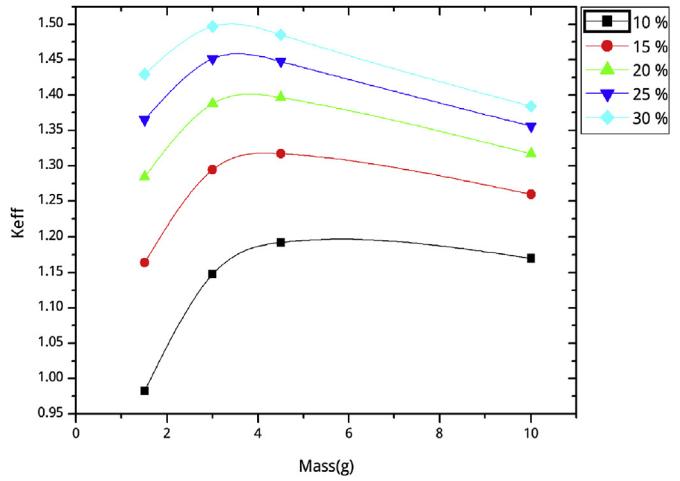


Fig. 2. Keff dependence on the radial reflector thickness.

Fig. 3. Keff dependence on the pebble fuel mass for different U²³⁵ enrichments.

A deep burn fuel cycle based on a mixture of 40% thorium and 60% uranium by weight was implemented (Talamo and Gudowski, 2005). The dependence of the multiplicative properties of the pebble's fuel mass on different fuel enrichment values was studied with the aim of obtaining the best fuel-moderator ratio and the lowest U²³⁵ enrichment, in a way that the critical conditions of the system would be ensured in a burning cycle as large as the standard LWRs'.

The K_{eff} values for the different proposed states were calculated with the MCNPX code. In Fig. 3, the calculations results for different pebble fuel masses (1.5 g, 3 g, 4.5 g, 10 g) and U²³⁵ enrichments by weight (10%, 15%, 20%, 25% and 30%) are shown.

For all U²³⁵ enrichment values, a maximum value for K_{eff} was obtained between 2 and 5 g of pebble fuel mass. A K_{eff} value of 1.1905 corresponding to a pebble fuel mass of 4.5 g with a 10% of U²³⁵ enrichment, was obtained, which is high enough to guarantee an adequate cycle's duration.

The latter demonstrates that relatively low U²³⁵ enrichment in a U + Th mixture is sufficient to obtain an excess reactivity high enough to reach an adequate cycle's duration in the critical system. In previous studies about U + Th fuel cycles, a 20% of U²³⁵ enrichment for a prismatic reactor was proposed (Talamo and Gudowski, 2005).

A homogeneous composition was considered for the inner part of each pebble (except for the coating), using the adequate mass fraction of carbon, silicon and fuel. It was considered that fuel particles are diluted in a carbon matrix and all the volume not occupied by fuel or by silicon carbide is occupied by carbon with a density of 1.796 g/cm³. For simulation proposes, the core was divided into ten horizontal levels considered in the calculation as homogeneous zones.

After optimizing the U²³⁵ enrichment and the mass per pebble values, radial and axial power density profiles and radial and axial

Table 3

Contribution of each fissile isotope to the total energy produced in both, the reactor and the ADSs.

Isotope	Reactor		ADSs	
	Released energy (MWd)	% of total	Released energy (MWd)	% of total
U ²³³	1402.8	3.5	1148.6	10.2
U ²³⁵	31485.2	78.7	7646.2	57.9
Pu ²³⁹	5582.8	14.0	3198.6	24.2
Pu ²⁴¹	281.7	3.8	1010.1	7.7
Total	40000	100	13200	100

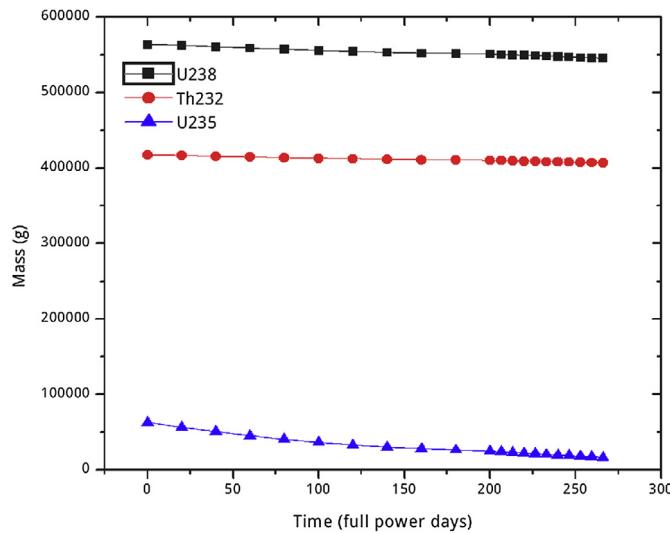


Fig. 4. Mass depletion of the initial fertile and fissile isotopes during both burning stages vs. the full power days.

neutronic flux distributions were obtained for the initial load and for the end of the cycle (EOC) in the VHTR core (not shown in this paper due to space constraints). Flux and power density profiles in the proposed VHTR present a similar behaviour to standard nuclear reactors. The peak factors are 1.25 and 1.16 for radial and axial distributions respectively. This shows uniformity of the power density profiles of the reactor core. After 200 days of fuel burning in the reactor core, fuel pebbles were redistributed into two ADSs cores with the same characteristics that the reactor (Table 2).

For the simulation of the fuel burning in both ADSs, 10 time steps of 11 days under a power of 60 MW_{th} were considered, ensuring to reach the design power at the EOC. The steady state was achieved after 20 sub cycles. Axial and radial power density profiles, as well as axial and radial neutron flux distributions at the beginning and the end of the stationary cycle, were also obtained for the two ADSs. The behaviour of the flux and power density in the ADSs is similar to the reference ADS.

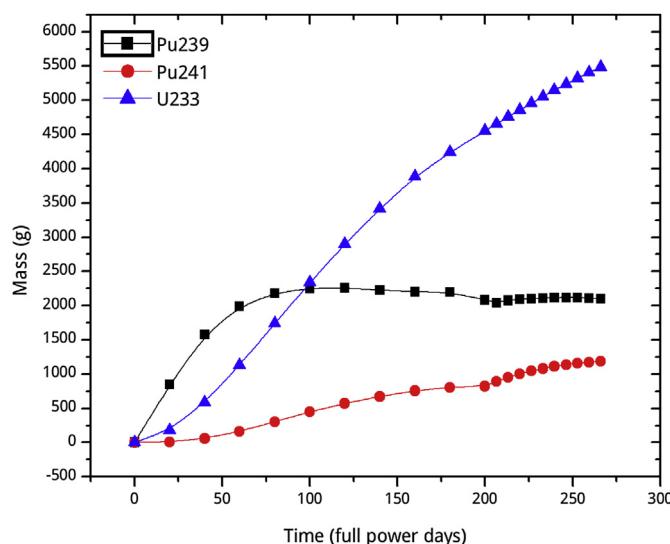


Fig. 5. Mass variation of the new fissile isotopes during both burning stages vs. the full power days.

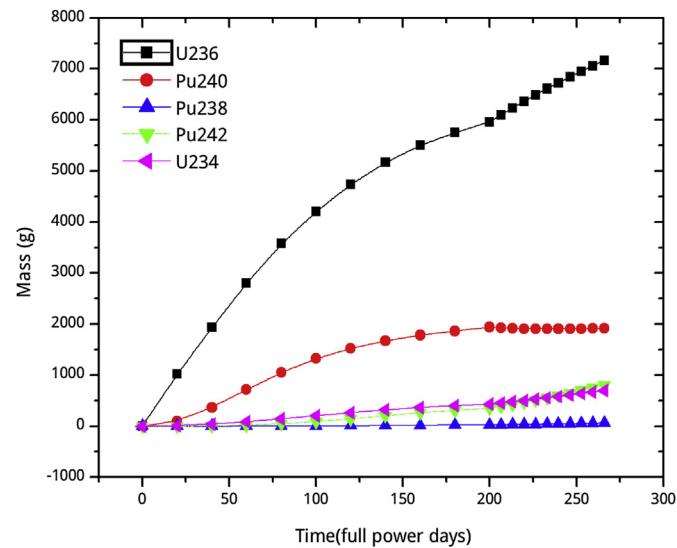


Fig. 6. Mass variation of uranium and plutonium isotopes during both burning stages vs. the full power days.

4.2. Fuel burnup

The reactor burn up was simulated; a mass of 4.5 g per pebble and a mixture of 60% of uranium and 40% of thorium by weight with a 10% of U²³⁵ enrichment were used. The latter guarantees a reactivity excess enough to maintain the criticality of the reactor during 200 days working under a power of 200 MW_{th}. The initial fuel mass loaded in the reactor was 1042 kg.

The fuel burnup in the reactor was calculated using Equation (1):

$$\text{Fuelburnup(GWd/ton)} = \frac{\text{Power(GW)} \cdot \text{Time(days)}}{\text{Fuelmass(ton)}} \quad (1)$$

In the critical system, the fuel reaches a burnup of 38.0 GWd/ton in a cycle of 200 days. The fuel burnup model in the ADSs considers that the fuel extracted from the lowest layer of the reactor is burned

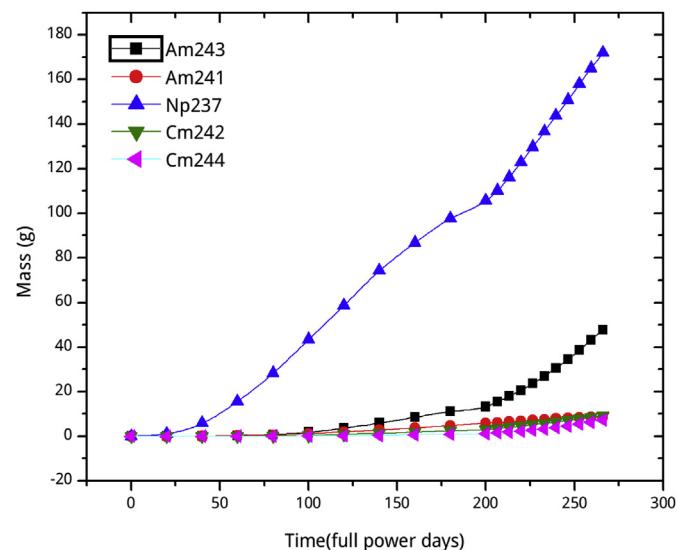


Fig. 7. Mass variation of minor actinides during both burning stages vs. the full power days.

Table 4

Fuel breeding coefficients in the U + Th cycle.

Stage	BC	%Fissile fuel
Reactor	0.44	52.1
Reactor + ADS	0.51	41.4

in the ADSs in a 110 days cycle under a power of 60 MW_{th}, ensuring that the subcritical system can reach that power at the end of each cycle in the steady state.

The burnup of the fuel extracted from the ADSs is 50.4 GWd/ton. This value is higher in more than 12 GWd/ton than the obtained in the critical system, and is also higher than the obtained in the conventional LWRs (40 GWd/ton), so the deep burn of the fuel is achieved.

One of the main goals of the proposed once-through U + Th cycle is the calculation of the contribution of the initial fissile isotope U²³⁵ and of those that originate from conversion, in this case Pu²³⁹, Pu²⁴¹ and U²³³, to the energy production, because the main goal of a deep burn once-through fuel cycle is to cause as much fission as possible of the initial fissile fuel and of the fissile isotopes obtained from transmutation of the fertile fuel.

The computational simulation of the proposed system using MCNPX allows calculating the average fission rate values for the considered fissile isotopes, at every burning step, and the final masses values of actinides. Taking into account the above data and the values of energy produced by fission, the contribution of each fissile isotope to the total produced energy was calculated. An analysis of each fissile isotope contribution to the total produced energy was done for the reactor and the ADSs.

The contribution of each fissile isotope to the total energy produced in the reactor and ADSs is shown in Table 3. It can be observed that the contribution of the plutonium fissile isotopes is greater than the contribution of U²³³.

In the second stage there is a greater contribution from the new fissile isotopes to the energy produced. Nevertheless, although there is a relative increase of U²³³ fissions, its contribution is still lower than the contribution of the set of Pu isotopes.

4.3. Variation of the isotopic composition

The mass variation of the main fuel isotopes for both burning stages is shown in Figs. 4–7. For the second burning stage, the sum of the isotope's masses from both ADSs is considered, and the effective time is considered as an equivalent time working under the reactor's power (200 MW_{th}), called full power days.

In Fig. 4 is observed a light depletion of fertile isotopes in relationship to fissile isotope. In Fig. 5 it can be seen that the mass of

Table 5

Mass (g) of plutonium isotopes and MA in a ton of spent fuel after 15 years of cooling in traditional LWRs (40.0 GWd/ton), in the VHTR (38.0 GWd/ton) and in the proposed hybrid system with a U + Th cycle (50.4 GWd/ton).

Isotope	LWR	Reactor	Reactor + ADSs
Pu ²⁴⁰	2600	1858	1844
Pu ²⁴¹	680	386	641
Pu ²⁴²	600	340	765
Pu ²³⁸	230	22	60
Pu ²³⁹	5900	2088	2167
Total	10010	4694	5437
Np ²³⁷	650	112	182
Am ²⁴¹	770	402	506
Am ²⁴³	140	13	45
Cm ²⁴⁴	31	1	5
Total	1591	528	738

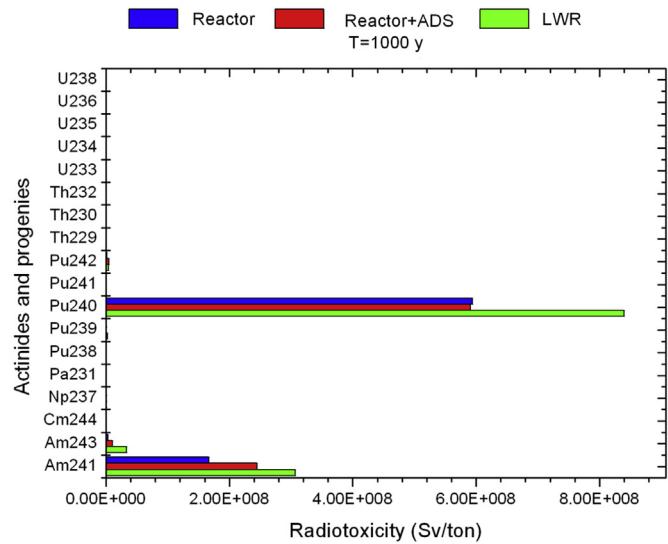


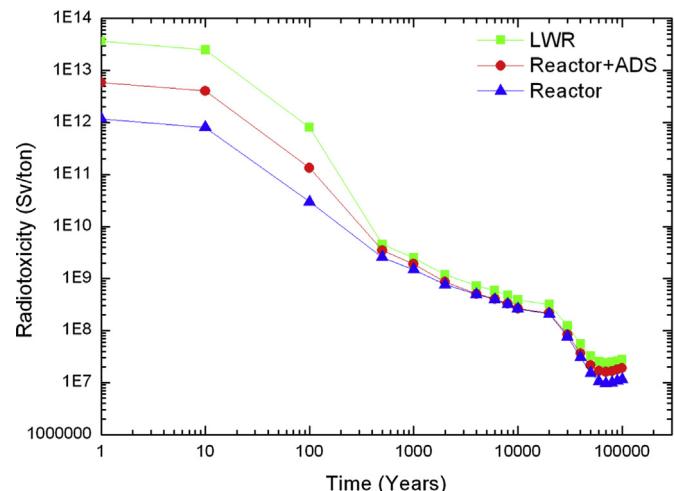
Fig. 8. Contributors to the inhalation radiotoxicity 1000 years after the fuel discharge.

U²³³ isotope presents a quasi linear growth during the first and second burning stages, in the reactor and the ADSs respectively. U²³³ isotope was obtained from transmutation of Th²³² isotope. However, Pu²³⁹ isotope reaches a stationary value in the reactor that remains during the burnup in the ADSs.

Fig. 6 shows that U²³⁶ isotope which is formed by neutron capture of the initial fissile isotope U²³⁵ and U²³⁴, increases faster in the second burning stage than in the first, but the mass of Pu²⁴⁰ isotope reaches a stationary value. The previous occurs because neutron capture rates of Pu²³⁹ and Pu²⁴⁰ are nearly equal in the second burning stage. Minor actinides which accumulate more are Np²³⁷ and Am²⁴³. Np²³⁷ is produced by neutron capture of U²³⁶ and subsequent beta disintegration of U²³⁷, and Am²⁴³ is produced from Pu²⁴² by the same process. In Fig. 7 is observed an increase in the accumulation rate of these isotopes during the second burning stage.

4.4. Fuel breeding coefficient (BC)

The fuel breeding coefficient (BC) was not calculated using the traditional expression that represents the quotient of the new

Fig. 9. Inhalation radiotoxicity provided by transuranic elements not including U²³³ and Pu²³⁹.

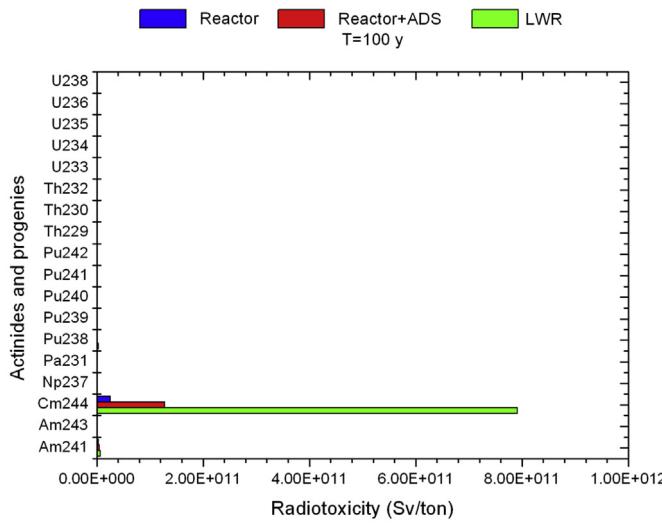


Fig. 10. Contributors to the inhalation radiotoxicity 100 years after the fuel discharge.

fissile isotopes production rate and initial fissile isotope consumption rate. It was calculated using the Equation (2) that expresses the relationship between the accumulated masses of new fissile isotopes and the depletion of initial fissile isotope. The above mentioned is equivalent to the original concept.

$$BC = \frac{Pu_{fis}^{239} + Pu_{fis}^{241} + U_{fis}^{233} + \Delta Pu^{239} + \Delta Pu^{241} + \Delta U^{233}}{\Delta U^{235}} \quad (2)$$

where:

Pu_{fis}^{239} , Pu_{fis}^{241} and U_{fis}^{233} . Mass of Pu^{239} , Pu^{241} and U^{233} that experienced fission.

ΔPu^{239} , ΔPu^{241} and ΔU^{233} . Mass of Pu^{239} , Pu^{241} and U^{233} accumulated at the end of the cycle.

ΔU^{235} . Spent mass of U^{235} .

The BC at the end of the cycle in the reactor, and at the end of the cycle including reactor and ADSs, are shown in Table 4. In both cases, the final amount of fissile fuel accumulated after 15 years of spent fuel cooling, is considered.

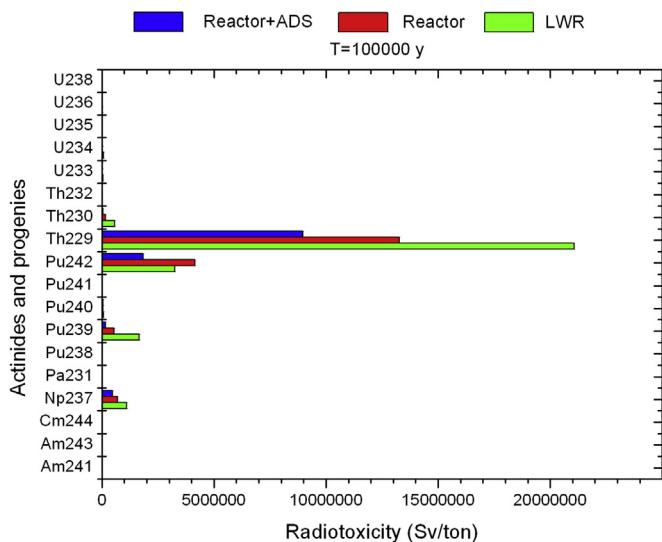


Fig. 11. Contributors to the inhalation radiotoxicity 100 000 years after the fuel discharge.

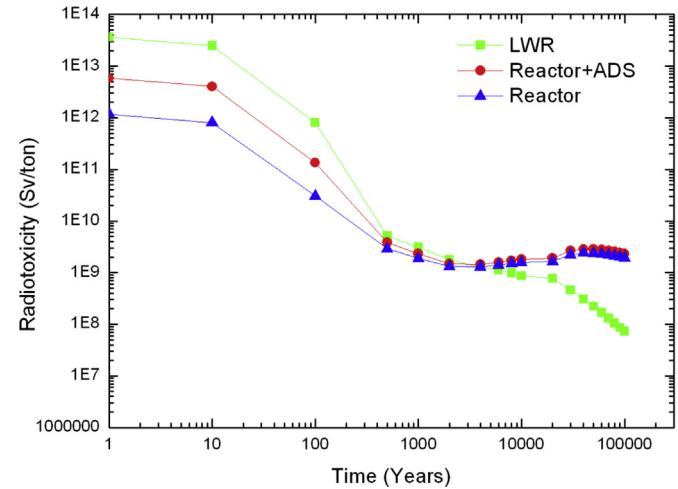


Fig. 12. Inhalation radiotoxicity provided by transuranic elements including U^{233} and Pu^{239} .

Extending the fuel cycle with the second stage in the two ADSs, a considerable increase of the fuel breeding coefficient is obtained, and therefore the utilization of the fertile isotopes U^{238} and Th^{232} is improved.

A parameter that allows evaluating the efficiency of a one-through cycle, in which the spent fuel is destined to long term storage, is the final percent of fissile fuel mass (%Fissile fuel).

$$\%Fissile\ fuel = \frac{Pu_{final}^{239} + Pu_{final}^{241} + U_{final}^{233} + U_{final}^{235}}{U_{initial}^{235}} \quad (3)$$

where:

Pu_{final}^{239} , Pu_{final}^{241} , U_{final}^{233} and U_{final}^{235} . Final mass of Pu^{239} , Pu^{241} , U^{233} and U^{235} .

U^{235} . Initial mass of U^{235} .

The final percent of fissile fuel that would be used in a cycle with fuel reprocessing is also given in Table 4.

Extending the fuel cycle with the second stage in the ADSs substantially reduces the mass of fissile fuel that would be sent to

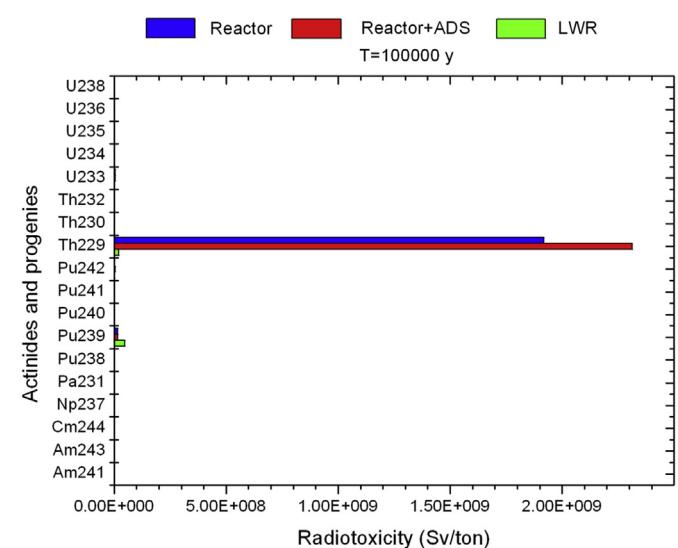


Fig. 13. Contributors to the inhalation radiotoxicity including U^{233} and Pu^{239} , 1,00,000 years after the fuel discharge.

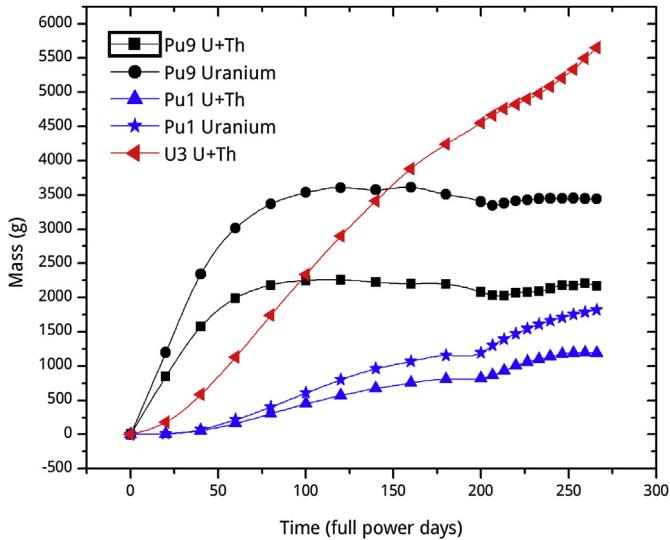


Fig. 14. Mass variation of the new fissile isotopes as function of full power days, for both burning stages in U and U + Th cycles.

long term storage if a once-through cycle were used. This is because an increase of the fuel burnup is obtained and although in the ADSs the mass of U^{233} continues to grow linearly, the mass of Pu^{239} reaches a stationary value and U^{235} decreases more intensely.

4.5. Radiotoxicity of the long lived wastes

The total mass of plutonium isotopes accumulated in a ton of wasted fuel in the proposed hybrid system (Reactor + ADSs, with a burnup of 50.4 GWd/ton), after 15 years of cooling, is 54.7% of the mass accumulated in a traditional fuel cycle of an LWR with a burnup of 40 GWd/ton (The European Technical Working Group on ADS, 2001), as shown in Table 5. If it is considered that 6429 g of U^{233} are also accumulated in the U + Th fuel cycle in the hybrid system, and that U^{233} is equivalent to Pu^{239} , the accumulated masses of plutonium isotopes in LWR cycle and the ones of plutonium isotopes plus U^{233} in the hybrid system are similar. However, the amount of MA accumulated in the proposed U + Th cycle for the hybrid system is much lower than in a traditional fuel cycle for an LWR, in spite of the notable difference in the achieved burnup values.

In the hybrid system cycle in relation to an LWR cycle, the 46.4% of masses of MA is accumulated. Also an important reduction of the accumulated masses of Am^{241} and Pu^{240} is obtained, which make an important contribution to the radiotoxicity of spent fuel during the first thousand years of decay (Fig. 8).

The increase of long lived wastes in the second stage in the ADSs is related to the increase of the burnup, and hence, to the additional amount of energy produced. Although the masses of Pu^{239} and Pu^{240} remain constant, there is a significant increase of the masses of Pu^{241} and Pu^{242} isotopes.

The dependence on time of the inhalation radiotoxicity provided by the transuranic elements (not including U^{233} and Pu^{239}) accumulated at the end of a standard LWR cycle (40 GWd/ton), at

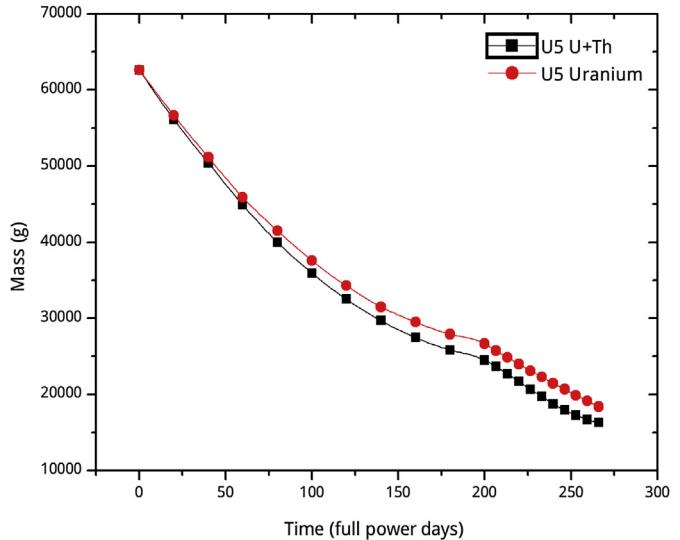


Fig. 15. Mass depletion of U^{235} as function of full power days, for both burning stages and for both analyzed cycles.

the end of the reactor cycle (38.0 GWd/ton) and at the end of the Reactor + ADSs cycle (50.4 GWd/ton), is shown in Fig. 9.

During the first 100 years the radiotoxicity is dominated by Cm^{244} and is higher in the spent fuel of the LWR, as can be seen in Fig. 10. At 1000 years, the radiotoxicity remains higher in the spent fuel of an LWR (Fig. 8) and is dominated by Pu^{240} and Am^{241} as stated above, which increases their mass in this type of cycle. At 100 000 years (Fig. 11), the main contributor to radiotoxicity is Th^{229} , descendant of U^{233} which is formed from beta disintegration of Pa^{233} , which in turn comes from alpha disintegration of Np^{237} .

In Fig. 9 can be seen that the radiotoxicity provided by the waste of the Reactor + ADSs cycle is always higher than the radiotoxicity provided by the waste accumulated at the end of the reactor cycle. This is logical as in the hybrid system the burnup is higher. Nevertheless, after the first 500 years there is not much difference in the radiotoxicity provided by both wastes.

The radiotoxicity provided by the long lived wastes including the new fissile isotopes accumulated at the end of the cycle (Pu^{239} and U^{233}) is shown in Fig. 12. After 5000 years the inhalation radiotoxicity in the U + Th cycle is higher than in a standard LWR cycle. This is due to accumulation of U^{233} in thorium-based cycles because it produces Th^{229} , which is the most contributor to the radiotoxicity between 10,000 and 100,000 years, as shown in Fig. 13.

4.6. Comparison between U + Th cycle and an equivalent uranium cycle

The U + Th fuel cycle composed of 40% of thorium and 60% of uranium with 10% of U^{235} enrichment was compared to a uranium

Table 7

Contribution (in percent) of each fissile isotope to the total energy produced in the reactor (first stage of the hybrid system) and in the ADSs (second stage of the hybrid system), for both cycles.

Stage	Cycle/Fissile isotope	U + Th	Uranium
Reactor	U^{233}	3.5%	0
	U^{235}	78.7%	76.4%
	Pu^{239}	14.0%	18.5%
	Pu^{241}	3.8%	5.1%
ADSs	U^{233}	10.2%	0
	U^{235}	57.9%	52.5%
	Pu^{239}	24.2%	36.0%
	Pu^{241}	7.7%	11.5%

Table 6

BC values and final percent of fissile fuel mass for a U + Th and uranium fuel cycles in the proposed hybrid system.

Parameters	Stage	U + Th	Uranium
BC	Reactor	0.44	0.38
	Reactor + ADSs	0.51	0.46
%Fissile fuel	Reactor	52.1	49.2
	Reactor + ADSs	41.4	36.8

Table 8

Mass (g) of the produced non fissile uranium and plutonium isotopes and MA, per ton of initial fuel in both cycles after 15 years of decay.

	U ²³⁶	U ²³⁴	Pu ²³⁸	Pu ²⁴⁰	Pu ²⁴²	Np ²³⁷	Am ²⁴¹	Am ²⁴³	Cm ²⁴⁴
Uranium	6580	0	56	2883	997	178	893	56	5
U + Th	6886	669	60	1844	765	182	506	45	5

fuel cycle with 6% enrichment, which is equivalent to the total enrichment of the previous mixture, in the proposed hybrid system. Both fuel cycles were subjected to the same power levels: 200 MW_{th} during 200 days in the reactor and 60 MW_{th} during 110 days in the ADSs. Obviously, the same burnup is reached in both cycle stages.

In Table 6, the values of BC calculated using Eq. (2) and the final percent of fissile fuel mass calculated using Eq. (3), are shown for both analyzed cycles. For both stages, a greater fuel breeding is obtained with the U + Th cycle; however, the final mass of fissile fuel left in the spent fuel is greater. This is due to the quasi linear increase of the U²³³ mass (Fig. 14), while the mass of Pu²³⁹ reaches stationary values since the first burning stage in the reactor, in both cycles.

Although the contribution of the new fissile isotopes to the energy produced is greater in the second stage than in the first stage of both cycles, it can be seen in Table 7 that this contribution is greater in the uranium cycle than in the U + Th cycle, i.e. a greater mass of Pu²³⁹ and Pu²⁴¹ experiment fission in the uranium cycle than the mass of U²³³, Pu²³⁹ and Pu²⁴¹ in the U + Th cycle. The previous corresponds to the larger depletion that undergoes U²³⁵ in the U + Th fuel cycle, shown in Fig. 15.

The previous comparison leads to the conclusion that the U + Th cycle is better than the equivalent uranium cycle in the proposed hybrid system, as long as the discharged fuel is reprocessed and the formed mass of U²³³ is utilized. In an open cycle, the traditional uranium fuel cycle is better.

The masses of non fissile uranium and plutonium isotopes and MA obtained per ton of initial fuel in both cycles after 15 years of decay are shown in Table 8. In the uranium cycle, the final masses of MA and Pu isotopes exceed those obtained in the U + Th cycle. However, in Fig. 16 it can be observed that for times longer than 2000 years, the inhalation radiotoxicity produced by the waste

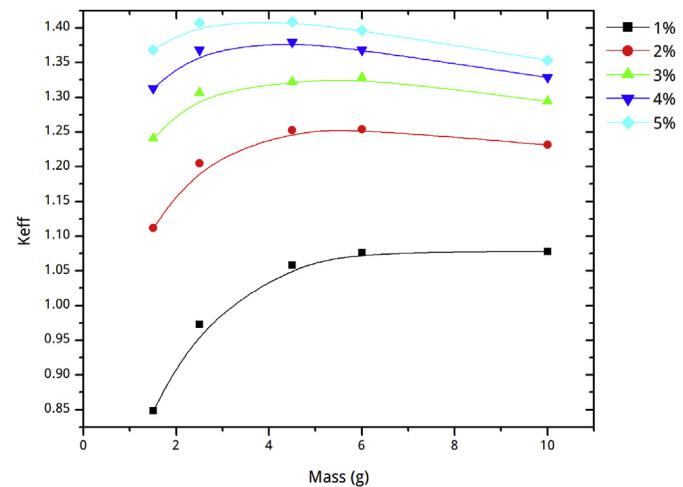


Fig. 17. Keff behaviour for different fuel masses per pebble and different Pu²³⁹ enrichments.

(including U²³³) of the U + Th cycle, exceed the inhalation radio-toxicity of the uranium cycle. This is due to the contribution of the Th²²⁹ isotope, which appears by disintegration of U²³³.

From the point of view of environmental impact of nuclear waste, the U + Th cycle has advantages with respect to the uranium cycle in the proposed hybrid system that uses the deep burn strategy, when using a fuel cycle with reprocessing, in which U²³³ produced from the Th²³² is returned to the nuclear reactors.

For a once-through and two stages cycle (with no reprocessing) in the proposed hybrid system, it is better to use the traditional enriched uranium fuel.

5. Plutonium + thorium cycle in the very high temperature hybrid system

A way to avoid the accumulation of plutonium formed during the reutilization of the fuel, i.e., the second generation plutonium, is to burn it combined with thorium instead of uranium, as Th + Pu-MOX. The ceramic fuel of VHTRs allows a high burnup which assures a high depletion of the initial loaded plutonium (86% of the

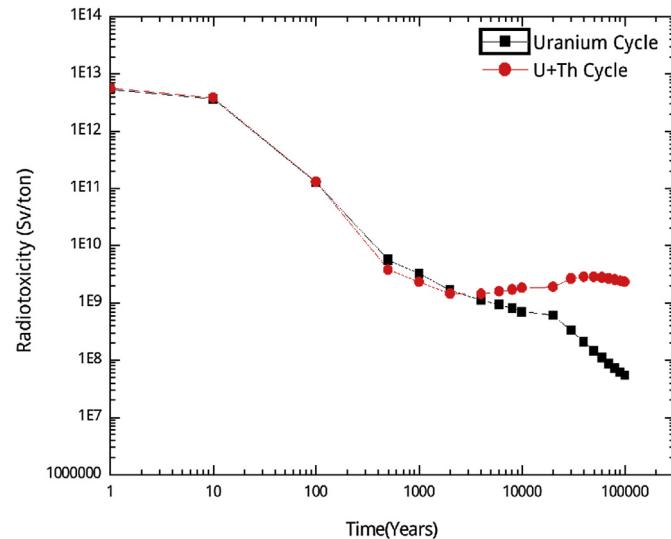


Fig. 16. Inhalation radiotoxicity provided by all actinides of the spent fuel at the end of both analyzed cycles in the hybrid system, as function of time.

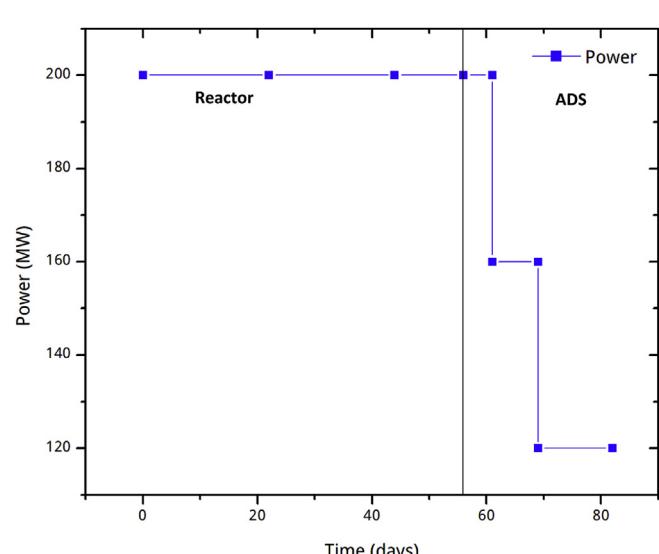


Fig. 18. Power vs. time for cycle 1 (Reactor + ADSs).

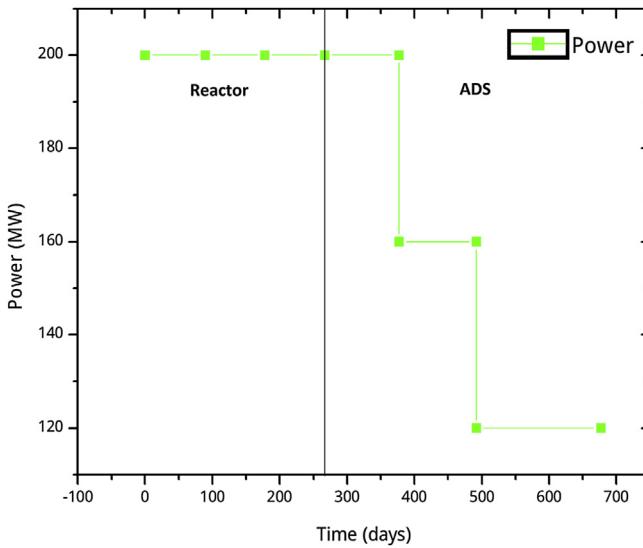


Fig. 19. Power vs. time for cycle 2 (Reactor + ADSs).

total plutonium approximately) when the first reutilization of plutonium takes place in a modular pebble-bed VHTR (Rutten and Hass, 2000).

The main goal of the plutonium-based fuel reutilization strategies is to burn as much plutonium as possible per unit of energy produced and minimize residual amounts of it in the repositories. In this section, a deep burn strategy for the utilization of the fuel mixture based on Pu^{239} and thorium (Th + Pu), in the proposed very high temperature hybrid system in a once-through and two-stages cycle, is studied. As it was considered before, when the fuel burned in the reactor cannot maintain the criticality of the system, the pebbles are moved to two ADSs and the critical system is refueled. The parameters of the reactor's and ADS's cores are shown in Tables 2 and 3 respectively.

In order to select the mass fraction of the metallic fuel components (Th^{232} and Pu^{239}) and the total fuel mass per pebble, the neutronic behaviour of the proposed reactor's core was modelled with fresh fuel, for different fuel masses per pebble and different proportions, which will represent the Pu^{239} enrichment. The mass

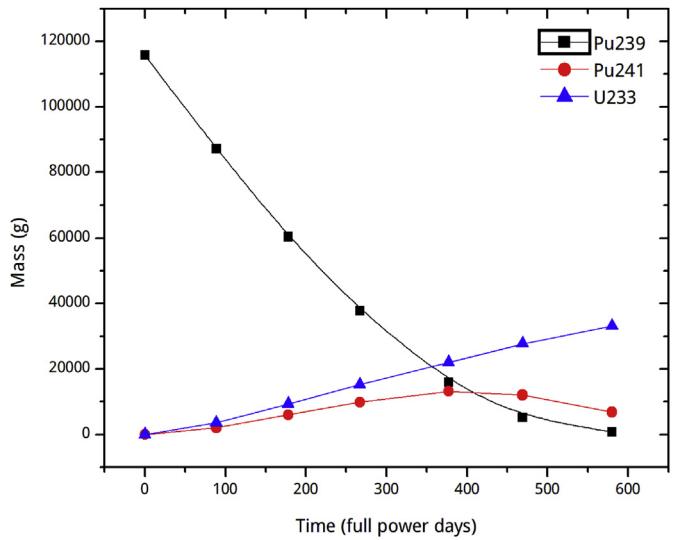


Fig. 21. Concentration of fissile isotopes, as function of time, for Pu + Th cycle 2.

values used were 1.5 g, 2.5 g, 4.5 g, 6 g and 10 g, and the Pu^{239} enrichments for each one of the masses were 1%, 2%, 3%, 4% and 5%. The results are shown in Fig. 17.

In Fig. 17 it can be observed that for Pu^{239} enrichment values equal to or higher than 2%, K_{eff} values higher than 1.20 are obtained if the fuel mass per pebble is greater than 4 g. In all cases, the dependence of K_{eff} on the fuel mass per pebble has a maximum between 2 and 6 g for Pu^{239} enrichments equal to or higher than 2%. For Pu^{239} enrichments equal to or higher than 3%, excess reactivity values remarkably high (around 30% or more) are obtained for most of the fuel mass values.

The following fuel compositions were chosen to evaluate the effectiveness of the Pu + Th fuel cycles in the proposed very high temperature hybrid system:

1. Fuel mass 4.5 g and 3% of Pu^{239} enrichment, hereinafter called cycle 1. The chosen mass is close to the optimum value corresponding to the chosen enrichment, which is an average value.
2. Fuel mass 10 g and 5% of Pu^{239} enrichment, hereinafter called cycle 2. The chosen mass and enrichment are extreme values in order to ensure the longest possible cycle and hence the higher fuel burnup.

Likewise the previous described U + Th cycle, a homogeneous composition was considered for the inner zone of each pebble. To simulate the continuous refuelling, the cores of the reactor and the ADSs were divided into 10 horizontal zones, and the same fuel management of the previous described cycle was carried out.

The variation of reactor and ADSs power, as function of time, for each of the proposed fuel cycles (1 and 2), is shown in Figs. 18 and 19.

In cycle 1 (Fig. 18), the fuel burning was modelled for the critical system under a power of 200 MW_{th}, and a cycle time of 56 days was

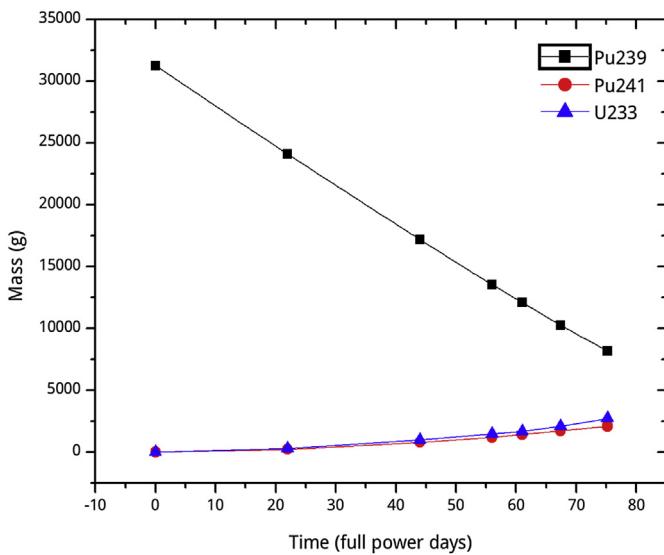


Fig. 20. Concentration of fissile isotopes, as function of time, for Pu + Th cycle 1.

Table 9

Values of BC, final fissile fuel masses in percent and depletion of Pu^{239} in percent, in the hybrid system (Reactor + ADSs), at the end of the studied cycles: Pu + Th cycle 1, Pu + Th cycle 2 and U + Th cycle. Asterisk (*) indicates that the value is referred to U^{235} .

Parameter	Pu + Th cycle 1	Pu + Th cycle 2	U + Th cycle
BC	0.34	0.73	0.51
% fissile fuel	48.8	35.6	41.4
% Pu^{239} depletion	26.2	0.7	39.1*

Table 10

Contribution of the fissile isotopes to the total energy produced in both stages of both Pu + Th cycles.

	Pu + Th cycle 1		Pu + Th cycle 2	
	Released energy (MWd)	%	Released energy (MWd)	%
Pu ²³⁹	14242	94.7	72122	62.2
U ²³³	170	1.1	19694	17.0
Pu ²⁴¹	628	4.2	24185	20.8
Total	15040	100	116000	100

Table 11

Mass (g) of Pu isotopes and MA per ton of spent fuel after 15 years of cooling: in traditional LWRs with a burnup of 40 GWd/ton, in the proposed hybrid system using the U + Th cycle with a burnup of 50.4 GWd/ton and using the Pu + Th cycle 2 with a burnup of 50.1 GWd/ton.

Isotope	LWR	U + Th	Pu + Th cycle 2
Pu ²⁴⁰	2600	1844	1950
Pu ²⁴¹	680	641	1450
Pu ²⁴²	600	765	2966
Pu ²³⁸	230	60	151
Pu ²³⁹	5900	2167	352
Total	10010	5517	6869
Np ²³⁷	650	182	22
Am ²⁴¹	770	506	1565
Am ²⁴³	140	45	319
Cm ²⁴⁴	31	5	41
Total	1591	738	1947

obtained. After the burning stage in the reactor, the fuel is moved to two ADSs and is burned under a power of 100, 80 and 60 MW_{th} during 26 days. In cycle 2 (Fig. 19), the fuel burning in the reactor lasts 267 days under the same power of cycle 1. Later in the ADSs the fuel is burned under a power of 100 MW_{th} during 110 days, 80 MW_{th} during 115 days and 60 MW_{th} during 185 days. The initial mass of the fuel loaded in the reactor is 1042 Kg for cycle 1 and 2315 Kg for cycle 2. The variation in the concentration of Pu²³⁹ and new fissile isotopes, as function of time (full power day), is shown in Figs. 20 and 21 for both studied cycles.

In Fig. 21 it can be observed that Pu²³⁹ is almost exhausted at the end of cycle 2, while the concentration of Pu²⁴¹ has a maximum value during the second stage, corresponding to the ADSs, and then

decreases; however, U²³³ experiences a linear increase. In cycle 1 (Fig. 20), a less depletion of Pu²³⁹ is reached, and the new fissile isotopes experience a much more pronounced growth in the second stage. This is related to the burnup reached in both cycles: in cycle 1, 10.8 GW/ton at the end of the first stage in the reactor and 14.4 GW/ton at the end of the whole cycle (Reactor + ADSs); in cycle 2, a deep fuel burnup is reached: 23.1 GW/ton in the first stage and 50.1 GW/ton at the end of the cycle, which is similar to the burnup obtained with the U + Th cycle analyzed in the previous section.

In order to compare the effectiveness of cycles 1 and 2, Three parameters (BC, %Fissile fuel and depletion of Pu²³⁹ in percent) were calculated and results are shown in Table 9. The BC and the final fissile fuel mass in percent were calculated using Eqs. (4) and (5) respectively. The depletion of Pu²³⁹ in percent was calculated as the ratio of its initial and final masses, multiplied by 100. The terms in the Eq. (4) and Eq. (5) have the same meaning as in Eq. (2) and Eq. (3).

$$BC = \frac{Pu_{fis}^{241} + U_{fis}^{233} + \Delta Pu^{241} + \Delta U^{233}}{\Delta Pu^{239}} \quad (4)$$

$$\% \text{Fissile fuel} = \frac{Pu_{final}^{239} + Pu_{final}^{241} + U_{final}^{233}}{Pu_{initial}^{239}} \quad (5)$$

In cycle 2, the proposed hybrid system reaches a good effectiveness in fuel utilization compared to cycle 1, because doubles the breeding of fissile fuel isotopes, and uses them to produce energy in the cycle itself. This is because the percent of fissile fuel in the waste is lower than in cycle 1 and in U + Th cycle. In addition, the initial Pu²³⁹ loaded in the reactor is almost completely exhausted at the end of cycle 2. Less than 1% of Pu²³⁹ initially loaded in the reactor is left as waste. In Table 10, the contribution of each fissile isotope to the energy produced in both stages for both Pu + Th cycles is shown. This information confirms the better utilization of the fissile fuel produced in cycle 2. A notable increase in the contribution of the new fissile isotopes to the energy production compared with cycle 1 is appreciated.

Taking into account the parameters shown in Table 9, it could be concluded that the Pu + Th cycle 2 is better than the U + Th cycle described in the previous section; both cycles reach similar total burnup values and cycle 2 reaches a greater cycle length (580 effective days instead of 266 days of the U + Th cycle). Nevertheless, although a great depletion of the initial Pu²³⁹ loaded as fissile fuel is obtained in cycle 2, a greater amount of plutonium isotopes per ton of initial fuel is produced.

In Table 11 it can be observed that the masses of Pu²⁴⁰, Pu²³⁸ and Np²³⁷ in a ton of the spent fuel of cycle 2, are lower than in a ton of the spent fuel of a standard LWR. However, the masses of Pu²⁴¹, Pu²⁴², Am²⁴¹, Am²⁴³ y Cm²⁴⁴ are higher. This difference can be compensated because the burnup in cycle 2 is higher than the burnup achieved in a standard LWR.

Comparing the sum of the masses of all transuranic waste isotopes per ton of initial fuel, taken from of Table 11, for the three cycles, arises the conclusion that the mass of the total waste in the standard LWR is the largest, with 11.6 Kg, followed by the mass of Pu + Th cycle 2 with 8.8 Kg, and finally the mass of the U + Th cycle with 6.2 Kg. Nevertheless, considering as waste the U²³³ produced, which occurs in a once-through cycle, the total mass of spent fuel would increase up to 24.6 Kg in cycle 2 and up to 12.6 Kg in the U + Th cycle. This reaffirms that thorium-based fuel cycles in a deep burn strategy are most useful if the remaining amount of U²³³ is retrieved by reprocessing the fuel.

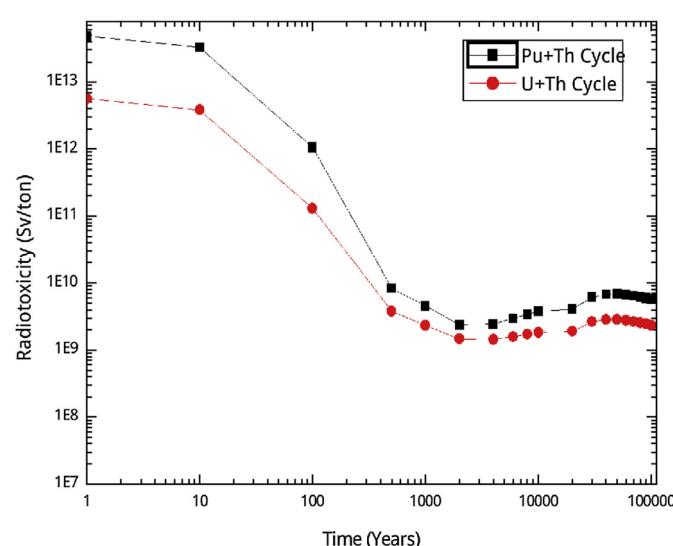


Fig. 22. Inhalation radiotoxicity as function of time, produced by all actinides of the spent fuel at the end of Pu + Th cycle 2 and U + Th cycle in the hybrid system.

In Fig. 22, the radiotoxicity as function of time, produced by all actinides of the spent fuel per ton of initial fuel for Pu + Th cycle 2 and for U + Th cycle, is shown. It is observed that the radiotoxicity of the waste of Pu + Th cycle exceeds that of the U + Th cycle, which has to do with the greater mass of the actinides in the Pu + Th cycle. From the point of view of environmental impact, the behaviour of radiotoxicity in Pu + Th cycle is unfavourable.

6. Conclusions

The preliminary conceptual design of a hybrid system composed of a graphite-gas pebbled bed very high temperature reactor and two ADSs of the same type was realized in order to analyze U + Th and Pu + Th fuel cycles, with the aim of obtaining a deep burn of the fuel. The cycles were divided into two stages: a first stage which takes place in the reactor and the second stage that takes place in two ADSs.

The extension of the fuel cycle by means of the second stage in the ADSs leads to an increase of the burnup and of the BC, and a decrease of the percent of fissile fuel in the spent fuel. Also, in the second stage of the U + Th cycle is obtained a greater contribution of the new fissile isotopes to the energy produced, than in the first stage.

A notable reduction of the Pu isotopes and MA stockpile is obtained in the U + Th cycle. The burnup achieved in the hybrid system (50.4 GWd/ton) is higher than the obtained with a standard cycle in an LWR (40 GWd/ton). However, if the U²³³ formed in the U + Th cycle is considered as waste, the radiotoxicity behaviour as function of time of the long lived wastes is worse than in a standard LWR's cycle.

The U + Th cycle in the proposed hybrid system was also compared with a uranium cycle with equivalent enrichment. With the U + Th cycle a greater fuel breeding is obtained compared to the uranium cycle, but the final amount of fissile fuel in the spent fuel is greater due to the increasing linear accumulation of U²³³. Therefore, if the U²³³ is recycled, the deep burn U + Th cycle in the hybrid system is more advantageous than the uranium cycle.

In the proposed hybrid system, two Pu + Th cycles were also studied. The cycle that uses 10 g of fuel mass per pebble and 5% of Pu²³⁹ enrichment (cycle 2) presents more advantages in achieving the deep burn of the fuel. This cycle reaches the same burnup that the U + Th cycle, almost the total exhaustion of Pu²³⁹, a greater BC and a lower percent of fissile fuel in the spent fuel discharged from the ADSs; nevertheless, a larger mass of Pu isotopes and MA is obtained in the discharged fuel. If it is considered again the U²³³ as waste, the final mass of long lived isotopes per ton of initial fuel in the discharged fuel exceeds that of the standard LWR's cycle, so its advantage lies in the reuse of U²³³. The simulations results for Th cycles show that the radiotoxicity of long term wastes increases for all proposed once-through cycles in relation to a standard LWR cycle, fundamentally for times longer than 3000 years.

7. Future works

The authors are trying to update the proposed hybrid system using as reference the Chinese 2 × 250 MWth HTR-PM demonstration plant (Z. Zhang et al., 2009).

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